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ACOUSTIC GAS RESONATORS FOR MEASUREMENT OF THERMOPHYSICAL PROPERTIES AND THERMOMETRY

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ABSTRACT

Gas-filled cavity resonators can be used to measure the speed of sound c, the viscous diffusivity D_v, and the thermal diffusivity D_t. The relative sensitivity of a complex resonance frequency to these parameters depends on the shape of the resonator and choice of mode. Different choices are appropriate for different measurements. Spherical acoustic resonators are excellent for very high-precision measurements of the speed of sound in dilute gases. The experimental models of spherical resonators include the effects of the thermal and viscous boundary layers, and the elastic deformations of the solid parts of the resonator. Spherical resonators have been applied to the measurement of the gas constant R, to acoustic thermometry, and to high precision measurements of the speed of sound $c(T,p)$ as a function of temperature and pressure for very pure gases. Precisions as high as a few parts in 10 million can be achieved. The ideal-gas specific heat and information about intermolecular interactions can then be extracted from $c(T,p)$. Cylindrical acoustic resonators are easier to fabricate and are suitable for many purposes, although they cannot be modeled as completely as spherical resonators. More complex resonator shapes have been developed for determination of D_v and D_t. The Greenspan acoustic viscometer, a resonator consisting of a cylindrical duct coupled at each end to large chambers, has a low-frequency mode in which the kinetic energy is localized in the duct, and the potential energy in the chambers. Its frequency response is strongly dependent upon D_v. As an absolute instrument, it is capable of determining D_v to a precision well below 1%. A cylindrical resonator with a honeycomb structure interposed in the flow midway between the ends has been developed for measurements of the ratio of D_v to D_t. The resonator shape is similar to the Greenspan viscometer, except that the chambers are coupled by a large number of parallel ducts. The modes of interest are similar to the plane-wave modes of a cylindrical resonator without the honeycomb insert. The odd plane wave modes interact with the insert mainly through viscous

coupling, and the even numbered modes interact with the insert primarily through thermal coupling. Measurements of the complex resonance frequencies of a set of odd and even modes can be used to determine the ratio D_v/D_t A recent addition to the models of acoustic resonators is the effects of surface roughness.

TRANSCRIPT

DR. MEHL: Thank you. I think the title makes it clear I am not an RUS person. I have for some 2 decades or so worked largely in collaboration with Mike Moldover and some other colleagues at NBS, now NIST, on a variety of acoustic and electromagnetic cavity resonators used to measure different properties of gases.

[Transparency 1]

I am going to talk today about the generic experiment and how we do some of our data analyses, pick out a few things that I think might be of interest to this community and this meeting.

Here is a generic experiment. We use a frequency synthesizer, generating a sine wave at discrete frequencies, controlled by a computer, a cavity resonator customized to the particular measurement, and a 2-phase lock-in amplifier. I think for many reasons it is important to measure both phases, the in-phase and quadrature signals in experiments of this type.

What I have plotted below is the absolute value of the complex signal u + iv at a set of discrete frequencies. This is a typical set of data. Here you see the resonance properties, the center frequency, f_0, and the half-width g. A Q would be the ratio of f_0:/(2g), which is the width here at $1/\sqrt{2}$ of the maximum.

[Transparency 2]

If you plot the same data in the complex plane, u versus v – at the right -- you see a circular plot for this ideal resonance. As you scan across the resonance, the points go around the circle.

This is quite useful in connection with some of the ideas that have been discussed here: How do you determine the Q of a resonance, and how do you determine the resonance frequency? Let's look at the theoretical expression here for the acoustic pressure in an enclosure. If I have a source at point r', the pressure at point r is given by this expression, which you can find in Morse and Ingard. There is a resonance denominator, there is another frequency up here.

If you parameterize this, you get an expression here for the microphone output, a complex signal. There is an amplitude A_N for each mode, and a complex resonance frequency for each mode.

Normally you are looking at a single mode of interest, but you will be sitting in the tails of many other resonances. It turns out that you can work with a small number of modes using the procedure I am going to talk about. Here I have isolated a single mode and then expanded the contributions from all the rest in a Taylor series.

This (B) is a complex number, a constant. The next term is a weak frequency dependence. The background terms take the origin of this complex product here and translate it in the complex u-v plane, so that the maximum distance from a point on the resonance to the origin need not correspond to the frequency f_0.

If my origin gets moved to, say, some point over here, then the amplitude is the distance from here to whatever the maximum distance along this curve is. That will, in general, will not be at f_0. You will get some distortion in the shape as you move around. It is clearly going to depend on where your background moves the origin to -- and I have exaggerated a bit here.

I think anyone who has looked at a lot of data will see a resonance that is sitting on a lot of background and you just see a little blip. Sometimes it looks like a bimodal signal. You can see why all this happens just by looking at this picture and thinking about how the distance from the origin varies as you sweep through the resonance, considering different levels of complex background.

Computers are very good at sorting this sort of thing out. I was telling someone earlier I did it for the first time in the early 1970s on a 8-bit lab computer with 1.8 K of memory. It requires a 6-parameter nonlinear least-squares fit. Today it is trivial and I think you could probably do it fast enough for a lot of modes in, say, an RUS experiment.

What we do is fit the theoretical function to our data set using the complex parameter A_N, the complex $F_N = f_0 + ig$, plus one or two complex background parameters, as necessary.

If you are looking at some closely spaced modes, you might include several terms with different mode indices N -- I have played around with up to 5, but 3 is trivial.

[Transparency 3]

What is more, this procedure is rather robust in determining f_0 and the half-width g, as shown by the algebra on this slide. Let's suppose that your transducer is measuring an acceleration instead of a velocity, or something in-between. Does that bias the measurement?

Suppose I put another factor of frequency f in the numerator. I can write that as my complex resonance frequency F + Δf, and separate off the constant term here, so I get a term that looks like the resonance term, except with a different amplitude.

Then the next term has a Δf in it, but the denominator is the product of $\Delta f = f - F_N$ and $f+F$, which is $2F+\Delta f$, something that is slowly varying, as the numerator is, so this second term really just modifies the background term.

More generally, any frequency dependence in this coefficient, which includes your transducer, response function, your amplifier, everything down the chain, just modifies the background. If you analyze data this way, you will get the resonance frequencies in spite of minor imperfections in your transducers.

[Transparency 4]

The ultimate limit is probably your detector, how good is the lock-in. This is an old slide. It comes from a paper by Moldover, Mehl, and Greenspan from the mid-1980s. Here are the 2 functions, u and v, and these are the residuals from a fit to a total of 22 measurements, 11 taken in rising frequency and then sweeping back down, so you see some reproducibility here.

The rms difference between the data and the fit is on the order of 0.02%. That means, in practice, that you can determine g, the half-width, and f_0, the resonance frequency, to about 0.02% of g, so the dissipation is determined to 0.02%, something on that order, and the frequency itself is determined with a fractional precision that is greater than that by the Q, often better than one part in 10^7.

The extent to which you could use this information depends on how well you can build a model of the experiment, that you can build in your understanding of the frequency. My point here is that there is plenty of resonance information from which to extract that information.

[Transparency 5]

This is from the same paper. It is from a mode that is nearly degenerate, nearly threefold degenerate. The lower plots show residuals. If you try to fit a 2-mode model to the data, you get something like 2% residuals, but with 3 modes the residuals go down to 0.02%, again, and you get good resonance parameters for all modes.

In this case there were 4 parameters for each resonance, so that is 12, total, plus at least one complex background, possibly 2, it says down here, so there would be 16 parameters in this fit. You span the range of nearly degenerate modes and you will pick them up.

[Transparency 6]

Okay, now some applications of this: For measurements of the speed of sound a good resonator is the sphere, which has some properties that make it suitable for talking about in this context of this meeting. I will spend a little bit of time talking about that example.

We have also made resonators that are good for measuring viscosity and thermal conductivity, properties dependent upon the measurement of the resonance widths. Here we are more interested in measuring the resonance frequencies of a spherical enclosure to determine the speed of sound, here written c (on some of my slides it will be u).

In the limit of 0 pressure in an ideal gas, c^2 equals the specific heat ratio, γ, times the gas constant R times the absolute temperature divided by the molecular mass.

At NIST, this in the mid-1980s, the group led by Moldover used the spherical resonator to measure R to 1.8 ppm, using argon and helium for which γ is exactly 5/3. (The molecular masses are well known and the experiment could be done at a known temperature).

More recently, the same apparatus has been used for primary acoustic thermometry between 217 and 303 K. It is possible to determine the absolute temperature scale to 0.6 mK. Measurements of this type make an important contribution to thermometry, where there are existing discrepancies among other measurements on the order of a few mK. There are plans to extend acoustic thermometry work up to 800 K at NIST.

[Transparency 7]

Now let's take a look at the modes of the sphere. One of the disadvantages of the spherical geometry is you cannot do anything about the spectrum; it is set by mathematics. The acoustic pressure will be proportional to eigenmodes. There is a spherical Bessel function, there is an eigenvalue, $Z_{ln,}$ and this is a spherical harmonic.

If I have a rigid sphere so that the acoustic velocity normal to the boundary must vanish at the boundary, then this Bessel function must have a vanishing derivative at the outer boundary. This is the boundary condition. The sphere also has to be thermally insulating; otherwise you do not have a pure acoustic wave inside.

In that limit this is the spectrum. There is a set of modes here at the bottom. There is a 0-frequency mode with l = 0. Here is the lowest-frequency breathing mode, f and a series of similar modes with radial motion.

Then there are modes with $l = 1$, which are all threefold degenerate, which you can deal with, but you would rather not, if you can avoid it, and then there are some messy ones up above. Sometimes there is a near degeneracy. Here is a particularly bad one within 0.2% of a radial mode here, which is, therefore, essentially unusable.

This mode has index $l = 13$. It is, therefore, 27-fold degenerate in a perfect sphere. The effects of imperfect geometry lift that degeneracy and broaden out a packet of modes that is very difficult to do anything with, not surprisingly.

In practice, you can deal with about five or six modes in the low-frequency range that are very nice. The sound is incident upon the boundary normally, and everything is calculable, in principle, using exact theories; you do not have to do approximations. This is really classical physics. The major solutions go back to Kirchhoff in 1868.

[Transparency 8]

The physics of the gas is described by the longitudinal component and transverse components of the Navier-Stokes equation, Fourier's law of heat flow, conservation of mass, and conservation of energy. Kirchhoff showed that you get a fourth order partial differential equation in the temperature from this, and this, and that that has 2 modes, an acoustic mode and a temperature mode with our usual acoustic propagation parameter here, and a thermal mode that is largely confined to the region near the boundary -- this thermal penetration length here, this mode dies out to $e^{-2\pi}$ in a wavelength δ_t.

But it leads to an important correction to the frequencies, because you have a transition from an adiabatic wave within the gas to an isothermal wave at the boundary, so some fraction of the volume proportional to delta t/a has an intermediate speed of sound, and this leads to a fractional correction that is proportional to $(\gamma-1)/2$ times δ_t,.

Shell motion: Some people in this room know that an isotropic solid with spherical shape is an exactly soluble problem. We have used classical elastodynamic theory to solve for the response of the shell to the internal pressure field, so that goes into the model as well.

[Transparency 9]

This slide summarizes the results of such a calculation. You have measured frequencies related to the speed of sound (u here), a mathematical eigenvalue, and the radius of the sphere.

The second term is the thermal boundary layer correction and this is the correction for shell motion. We can skip the width expressions here.

For the sphere used for measuring the gas constant, the thermal boundary layer correction is 0.17% for the lowest mode at a pressure of 1 bar and scales with the reduced pressure and frequency variables, as shown here. The shell correction is 1.7 ppm at 1 bar and it has a resonant denominator, which I will say a little bit more about.

[Transparency 10]

The other thing we have to worry about is the effect of imperfect geometry, and a sphere is nice for this. If you consider a deformed sphere, where a parameter ε sets the scale and this function, curly F is of order unity, you can show that if you pick an F that preserves the volume, then the eigenvalue spectrum for the radial modes with quantum number $l = 0$ is perturbed in order ε^2.

You can make a sphere in a good machine shop with ε on the order of a few times 10^{-4}, so you get down to corrections that are generally less than a part in 10^6 for the radial modes.

The nonradial modes, beside being degenerate, are all perturbed in order ε. This is a potential problem, because you have to measure this mean radius somehow. The only good way to do it as a function of temperature is to use the electromagnetic modes for which there is no non-degenerate s-wave-type mode. It turns out that if you consider any multiplet, do an average over the geometric perturbation, the average frequency of any multiplet is perturbed in order ε^0 -- this applies to the acoustic modes, to the electromagnetic modes of the electrical mode class, and the magnetic mode class, 3 different boundary conditions, and I would be willing to bet it also applies to solid elastic spheres.

[Transparency 11]

A little bit more about this elastic correction. I want to show you some evidence that we have for this, and then I will have to stop; I will not be able to say anything about the other experiments.

The equation at the bottom shows the effects of shell motion for different symmetries. There is a shell-admittance function, dependent on frequency and the mode index l. Fractional corrections have a coupling constant. In the numerator, ρc^2 for the gas is essentially γ times the ambient pressure, and in the denominator ρc^2 for the solid is an elastic constant. For gases this

ratio is a relatively small number and that is the source of the low magnitude -- of order 1.7 ppm, typically.

The shell admittance function is plotted at the top. For the l = 0 you see the resonance here (for the 3-l sphere that was around 14 kHz). For l = 1 there is a 0-frequency resonance that corresponds to translational motion and another resonance over here. Here is a bending-mode resonance for l = 2 symmetry.

[Transparency 12]

These data are from an earlier, thinner, sphere. They show the shell perturbation for a series of modes. There is evidence here of the singularity at the shell breathing mode, a change in the sign of the correction as you pass through the mode.

[Transparency 13]

This shows something similar for the l = 1 triplets in that same geometry. Generally there are 3 points at each frequency, all generally falling along the same curve, and we just pick up a hint of the singularity here. But look at what happens to the lowest mode; it is going way up, up the graph and you see a separation in the 3 components of the multiplet. These modes have the gas oscillating in these directions.

The sphere is suspended, so there is a little bit of stiffness for this (vertical) motion, greater stiffness than for these 2 (horizontal) motions. The motions in the horizontal plane have about the same fractional correction and the vertical one is a little bit smaller. In fact, if you make the stiffness in the vertical direction larger and larger, you can force that correction down and even change its sign.

These experiments show us something about the coupling of the fluid inside to the mechanical motion of a shell and it makes you speculate a little bit about what can be done. Can you make the shell stiff enough so it does not matter so much?

[Transparency 14]

The answer is displayed here on this slide, where I show the resonances of a spherical shell as a function of the ratio of the outer to the inner radius. Here is the breathing mode. Its frequency decreases with increasing wall thickness. Of course, the corresponding correction gets larger at low frequencies as it gets stiffer, but you cannot make the resonance go away. You make the shell a little stiffer but you also increase the mass. The same is true for the l = 1 and l = 2 modes.

At the right some argon acoustic resonance frequencies are listed. There are generally several that fall below the extensional elastic modes here, so there is a little bit of room for doing an acoustic experiment in a gas inside of such an enclosure. If the gas is helium, there are not many modes below the shell breathing mode. If you start putting liquids in the shell, it is a real challenge. (I think some people have also found some clever solutions, too).

DR. SMITH: Have there been attempts to measure the __________ fluid by putting it inside a metal shell __________? Has that been your experience?

DR. MEHL: I have not tried to use this for liquids. We have worked with gases up to about 10 bar. The correction is 1.7 ppm. at 1 bar and only 17 ppm at 10 bar. You are up at least 2 more orders of magnitude. It should be possible if you can sort out the modes.

I would build a model that has mode coupling if I were doing such an experiment.

DR. SMITH: We did a 2-layer computation from a __________ shell. ___________ they were just Rayleigh waves in the metal.

DR. MIGLIORI: Last week I talked with Mike Moldover. He had attempted to measure the elastic constants of a batch of Invar that he was using for spherical shells and he failed because the microstructure of that Invar was so bad he could not get anything close to a fit for the elastic moduli, but it appears that he needed to know the elastic moduli of the shell about 5 times better than we had any hope of ever measuring, partly because it was Invar, partly because of constraints.

Have things gotten better?

DR. MEHL: He is probably hoping that he can build an apparatus that will work up to 800 K. For the stainless steel sphere here, the breathing mode compliance was measured in situ by filling the sphere with pressurized mercury.

DR. MIGLIORI: Stainless steel is much nicer than Invar.

DR. IZAAK: You said the container needs to be thermally insulated, otherwise there would be no acoustic wave inside?

DR. MEHL: No. In a gas there is strong coupling between the pressure and temperature fields. You have an adiabatic wave. As pressure oscillates, the temperature oscillates -- in dimensionless units they oscillate about the same amount. Any solid boundary is a very good thermal conductor compared with the gas.

There will be a heat flow, an oscillating heat flow, in the boundary region, whose thickness is given by this δ_t. When I was showing you the eigenvalues in the absence of such approximation, that corresponds to the limit of an insulating shell.

DR. MARSTON: Is it common to go back and check whether the background that you introduce for any one mode, say, the local background near your mode, is consistent with the strengths you have from the adjacent modes?

DR. MEHL: I played around with that a little bit years ago. We have never pursued it very seriously, no. But it is certainly qualitatively true in that if you are working with a high mode, you are sitting on the shoulders of a lot of other resonances and you would see a high background, whereas in the lower part of the spectrum you see small background.

What we typically do is make sure that the fits are insensitive to this, that is, try adding another parameter and seeing if it makes a difference and reject data that do not fit sensible behavior patterns.

I should also mention, since I said that we use the measurements of the half-width to determine dissipative processes, that it is important to refine the models. For our acoustic viscometer we take into account the fact that the dissipation, which is viscous itself, is proportional to the square root of the frequency.

You build a resonator to measure the width, you do not want it to have such a high Q -- 100 or so is fine. When you build a model for that, it is important to put the frequency dependence of the width into the model, do not assume it is a constant. We actually do a little better than that, but that is the minimum you should do.

Thank you.